Photoinduced magnetization of a gas of atoms with hyperfine structure

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We report the first investigation of the photomagnetization of a gas containing active atoms with hyperfine structure due to the interaction of the electronic subsystem with the nuclear spin. Perturbation calculations are performed for the nonstationary and stationary regimes as well as in the case of the passage of an ultrashort pulse. Spontaneous photon emission in the excited states of an atom is taken into account. This leads to optical pumping with the atom being transferred from a resonant sublevel of the ground state to all sublevels of the ground state that are allowed by the selection rules for the total angular momentum. We find a nonlinear effect of optical pumping in the photoinduced magnetization of the gas. This effect influences the magnitude and direction of the magnetization. It is shown that the hyperfine structure of the atomic levels gives rise to characteristic hyperfine structure in the photoinduced magnetization of the gas. The behavior obtained are illustrated by specific examples using a rubidium atom with doublet splitting of the ground and excited states. © *1995 American Institute of Physics.*

1. INTRODUCTION

Experiments on the nonlinear magnetization of solids[1–4] and atomic gases[5–8] by a light wave are conducted both to solve fundamental problems of the interaction of light with matter and to determine the relaxation constants. Both a phenomenological approach[6–8] and a rigorous quantum- mechanical analysis based on the density matrix for atoms with zero nuclear spin in the absence[9] or presence[10] of an external magnetic field have been used to explain this magnetooptic phenomenon in atomic gases. However, experiments with atomic gases usually employ atoms that have nonzero nuclear spin,[5–8] in which case optical pumping populates not only resonant hyperfine components of the levels, but nonresonant ones as well. This significantly alters the magnetic moment per unit volume of the gas, a phenomenon known as photoinduced magnetization. In this connection it is of interest to construct a detailed theory of photoinduced magnetization of a gas of atoms possessing hyperfine structure.

In the present paper the photoinduced magnetization of such a gas is investigated theoretically using the equations for the density matrix, taking into account the degeneracy of the sublevels with respect to the projection of the total angular momentum, thermal motion, irreversible relaxation, and the arrival of atoms in a sublevel of the lower state as a result of spontaneous emission of a photon in the excited state. The latter process leads to optical pumping, with the atom being transferred from a resonant sublevel of the ground state E_a to all sublevels of the ground state allowed by the selection rules for the total angular momentum. This results in a new nonlinear phenomenon—optical pumping in photoinduced magnetization. In this effect, at an exact resonance between the hyperfine sublevels of the ground E_a and excited E_b states, both the aforementioned resonant sublevels and the nonresonant sublevels of the ground state E_a that are filled as a result of optical pumping contribute to the photoinduced magnetization. As a consequence, the photoinduced magnetization acquires characteristic hyperfine structure in the form of individual bell-shaped peaks when either the frequency of a cw monochromatic source or the center frequency of an ultrashort pulse is swept, inducing both nonstationary and constant asymptotic magnetizaton. These peaks incorporate contributions to the photoinduced magnetization from individual hyperfine sublevels of the ground level E_a and the excited state E_b. Depending on the separation between sublevels, Doppler width, and relaxation constants, some of these peaks can merge, and the resultant can be observed by sweeping the frequency of the transmitted light wave.

Optical pumping significantly influences the magnitude and direction of photoinduced magnetization of an individual peak, since the sublevels filled by optical pumping have differing total angular momenta, which strongly affect the magnetization of an atom in a given sublevel. When the indicated peaks are completely separated, the separation of the corresponding hyperfine components in both the ground state E_a and the excited state E_b can be determined experimentally by measuring the distance between maxima. The optical pumping effect also shows up prominently in the behavior of the photoinduced magnetization as a function of time t' during the passage of an ultrashort pulse with fixed center frequency and duration τ, if irreversible relaxation characterized by the constants γ_a and γ_b in the ground E_a and excited E_b states, where $(\gamma_a+\gamma_b)\tau \ll 2$, occurs in the gas. In this case, by virtue of resonant interaction with the ultrashort pulse, the photoinduced magnetization peaks at $t'_{m_1} \approx \tau$, and after the passage of an ultrashort pulse with $\tau < t'$ this magnetization decays by an irreversible relaxation mechanism over a time γ_a^{-1} or γ_b^{-1}.

In addition, because the excited state of the atom decays spontaneously, optical pumping comes into play and fills one resonant and several nonresonant sublevels of the ground state E_a, and this leads to an alternative atomic magnetization mechanism with $\tau < t'$, due to the optical pumping ef-

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fect. This photoinduced magnetization based on optical pumping changes from zero to some maximum (minimum) value at time

$$t'_{m2}=(\gamma_b-\gamma_a)^{-1}\ln(\gamma_b/\gamma_a)>t'_{m1},$$

after which it decreases (increases) and affects both the magnitude and direction of the total photoinduced magnetization, incorporating all possible contributions. An experimental investigation of the rate of decay of the total photoinduced magnetization with $\tau<t'$ makes it possible in principle to determine the relaxation constants γ_a and γ_b.

The physics of the nonlinear optical pumping effect in the photoinduced magnetization is simple, and it can be easily observed experimentally.

2. BASIC EQUATIONS FOR ATOMS WITH NUCLEAR SPIN

We now consider a nonmagnetic atomic gas containing active atoms with nuclear spin, as occurs in a number of experiments.[5–8] In the presence of nuclear spin, the energy level E of an atom is split into a series of hyperfine components with energies[11]

$$E_F=E+\hbar\Delta_F,\quad \hbar\Delta_F=AC/2+BC(C+1),$$

$$C=F(F+1)-J(J+1)-I(I+1),$$

where E is the energy of the atom neglecting nuclear spin, $\hbar\Delta_F$ is the hyperfine interaction energy, and A and B are the magnetic and quadrupole splitting constants of the level; F, J, and I are the quantum numbers of the total angular momentum $\mathbf{F}=\mathbf{J}+\mathbf{I}$, the electronic angular momentum $\mathbf{J}$, and the nuclear spin $\mathbf{I}$. The state of an atom in a particular hyperfine sublevel E_F is characterized by the quantum numbers J, I, and F, and the projection $\hbar M_F$ of the total angular momentum on the quantization axis. For $J\geqslant I$ the level E is split into $2I+1$ hyperfine components, while for $J<I$ it is split into $2J+1$ components.

A plane light wave with a rotating electric-field vector $\mathbf{E}$

$$\mathbf{E}=\mathbf{l}_{\mathbf{k}\lambda}a(t')\exp[i(\mathbf{kr}-\omega t)]+\text{c.c.},\qquad(1)$$

where

$$t'=t-t_0-\mathbf{k}(\mathbf{r}-\mathbf{r}_0)/\omega,$$

propagates in such a gas. Here $\mathbf{l}_{\mathbf{k}\lambda}$ is a complex unit vector of elliptical polarization with indices $\lambda=1$ and $\lambda=-1$, respectively, for right- and left-hand rotation of the electric field, and $a(t')$ is the complex amplitude, which varies slowly compared with $\exp[i(\mathbf{k}\cdot\mathbf{r}-\omega t)]$; t_0 is the initial time at which the light wave (1) enters the atomic gas at the boundary point $\mathbf{r}_0$ of some volume containing active atoms. The frequency ω is close to the frequency $\omega_{F_bF_a}=(E_{F_b}-E_{F_a})\hbar^{-1}$ of the atomic transition between the hyperfine components

$$E_{F_a}=E_a+\hbar\Delta_{F_a}\quad\text{and}\quad E_{F_b}=E_b+\hbar\Delta_{F_b}$$

of the ground and excited levels E_a and E_b. Subscripts a and b denote physical quantities associated with the atom's ground state (E_a, J_a, F_a, M_a) and excited state (E_b, J_b, F_b, M_b). In general, the frequency ω and wave vector $\mathbf{k}$ satisfy the dispersion relation $\omega^2\varepsilon(\omega)=k^2c^2$, where c is the speed of light in vacuum and $\varepsilon(\omega)$ is the real dielectric constant,[12] which takes account of both resonant levels of the active atoms, and nonresonant levels of the active and impurity atoms in the isotropic gas under study.

We write the elliptical polarization vector $\mathbf{l}_{\mathbf{k}\lambda}$ in a general form that does not depend on the choice of coordinate system. For right-hand polarization $\lambda=1$, we have

$$\mathbf{l}_{\mathbf{k}1}=\mathbf{l}_{\mathbf{k}}^{(1)}\cos\psi+i\mathbf{l}_{\mathbf{k}}^{(2)}\sin\psi.\qquad(2)$$

For left-hand polarization $\lambda=-1$, the elliptical polarization vector is given by a different formula,

$$\mathbf{l}_{\mathbf{k},-1}=-\mathbf{l}_{\mathbf{k}}^{(1)}\sin\psi+i\mathbf{l}_{\mathbf{k}}^{(2)}\cos\psi.\qquad(3)$$

Here the argument ψ assumes values $0\leqslant\psi\leqslant\pi/2$. The quantities $\cos\psi/\sin\psi$ in Eq. (2) and $\sin\psi/\cos\psi$ in Eq. (3) characterize the ratio of the axes of the polarization ellipse. If $\psi=\pi/4$, the polarization ellipse becomes a circle, and Eqs. (2) and (3) refer, respectively, to right- and left-hand circularly polarized waves. If $\psi=0$ or $\psi=\pi/2$, the polarization ellipse deforms into orthogonal line segments, and Eqs. (1)–(3) describe linearly polarized waves. The unit vectors in Eqs. (2) and (3) satisfy

$$\mathbf{kl}_{\mathbf{k}}^{(1)}=\mathbf{kl}_{\mathbf{k}}^{(2)}=\mathbf{l}_{\mathbf{k}}^{(1)}\mathbf{l}_{\mathbf{k}}^{(2)}=0,\quad \mathbf{l}_{-\mathbf{k}}^{(1)}=\mathbf{l}_{\mathbf{k}}^{(1)},\quad \mathbf{l}_{-\mathbf{k}}^{(2)}=-\mathbf{l}_{\mathbf{k}}^{(2)},$$

$$\mathbf{l}_{-\mathbf{k}\lambda}=\mathbf{l}_{\mathbf{k}\lambda}^{*},\quad \mathbf{l}_{\mathbf{k}\lambda}\mathbf{l}_{\mathbf{k}\lambda'}^{*}=\delta_{\lambda\lambda'},\quad [\mathbf{l}_{\mathbf{k}}^{(1)}\mathbf{l}_{\mathbf{k}}^{(2)}]=\beta\mathbf{k}/k,$$

$$i[\mathbf{l}_{\mathbf{k}\lambda}\mathbf{l}_{\mathbf{k}\lambda}^{*}]=(\mathbf{k}/k)\lambda\beta\sin(2\psi),\quad \beta=(\mathbf{k}/k)[\mathbf{l}_{\mathbf{k}}^{(1)}\mathbf{l}_{\mathbf{k}}^{(2)}],$$

where β is a unit pseudoscalar.

In the resonance approximation, for which $|\omega-\omega_{F_bF_a}|\ll\omega$, the interaction of active atoms with the light wave (1) is described by means of the quantum-mechanical equations for the matrix elements of the density matrix ρ in the FM_F representation:

$$\left(\frac{\partial}{\partial t}+\mathbf{v}\nabla+i\omega_{F_bF_a}+\gamma_{ba}\right)\rho_{F_bM_{F_b},F_aM_{F_a}}$$

$$=\frac{i}{\hbar}(\mathbf{Ed}_{F_bM_{F_b},F'_aM_{F'}}\rho_{F'_aM_{F'},F_aM_{F_a}}$$

$$-\rho_{F_bM_{F_b},F'_aM_{F'}}\mathbf{Ed}_{F'_aM_{F'},F_aM_{F_a}}),\qquad(4)$$

$$\left(\frac{\partial}{\partial t}+\mathbf{v}\nabla+i\omega_{F_bF'_b}+\gamma_b\right)\rho_{F_bM_{F_b},F'_bM'_{F'}}$$

$$=\frac{i}{\hbar}\left(\mathbf{Ed}_{F_bM_{F_b},F_aM_{F_a}}\rho_{F_aM_{F_a},F'_bM'_{F'}}\right.$$

$$\left.-\rho_{F_bM_{F_b},F_aM_{F_a}}\mathbf{Ed}_{F_aM_{F_a},F'_bM'_{F'}}\right),\qquad(5)$$

$$\left(\frac{\partial}{\partial t}+\mathbf{v}\nabla+i\omega_{F_aF'_a}+\gamma_a\right)\rho_{F_aM_{F_a},F'_aM'_{F'}}$$

$$=\frac{\gamma_aNf(v)}{(2J_a+1)(2I+1)}\delta_{F_aF'_a}\delta_{M_{F_a}M'_{F_a}}$$

$$+\frac{i}{\hbar}(\mathbf{Ed}_{F_aM_{F_a},F_bM_{F_b}}\rho_{F_bM_{F_b},F'_aM'_{F'}}$$

$$-\rho_{F_aM_{F_a},F_bM_{F_b}}\mathbf{E}\mathbf{d}_{F_bM_{F_b},F'_aM'_{F'_a}})$$

$$+\frac{\gamma(2J_b+1)}{|d_{ba}|^2}\mathbf{d}_{F_aM_{F_a},F_bM_{F_b}}\rho_{F_bM_{F_b},F'_bM_{F'_b}}\mathbf{d}_{F'_bM_{F'_b},F'_aM'_{F'_a}}, \tag{6}$$

where

$$\omega_{F_bF_a}=(E_{F_b}-E_{F_a})\hbar^{-1}=\omega_{ba}+\Delta_{F_b}-\Delta_{F_a},$$

$$\omega_{ba}=(E_b-E_a)\hbar^{-1},\quad \gamma_{ba}=\frac{\gamma_b+\gamma_a}{2},$$

$$\gamma=\frac{4|d_{ba}|^2\omega_{ba}^3}{3\hbar c^3(2J_b+1)},$$

$$f(v)=(\sqrt{\pi}u)^{-3}\exp\left(-\frac{v^2}{u^2}\right),\quad u=\sqrt{\frac{2k_BT}{m_{at}}},$$

$\mathbf{d}_{F_bM_{F_b},F_aM_{F_a}}$ is the matrix element of the electric dipole moment operator $\mathbf{d}$, γ_{ba} is the half-width of the resonance transition spectral line, $\hbar\gamma_a$ and $\hbar\gamma_b$ are the homogeneous widths of the ground state E_a and the excited state E_b, γ is the probability of a spontaneous emission of the photon $\hbar\omega_{ba}$ by an isolated atom, d_{ba} is the reduced dipole moment of the atomic transition $J_a\to J_b$,[13] $f(v)$ is the Maxwell distribution, $\mathbf{v}$ is the velocity of the atom, u is the most probable velocity, $\varkappa_B$ is Boltzmann's constant, T is the temperature, m_{at} is the mass of an atom, and N is the stationary density of atoms in the ground state E_a, taking into account all components of the hyperfine structure with $\mathbf{E}=0$. The magnitude of γ_a, γ_b, and γ_{ba} is determined by the radiative decay and inelastic atomic collisions. The term in Eq. (6) containing γ describes the arrival of atoms in the ground state E_a as a result of spontaneous emission in the excited state E_b. The term in Eq. (6) containing the product $Nf(v)$ takes into account the Boltzmann distribution of the atoms over the levels and the Maxwell–Boltzmann velocity distribution in statistical equilibrium prior to the passage of the light wave (1). Since $\hbar\omega_{F_bF_a}\gg\varkappa_BT$, the excited state E_b is unpopulated before the interaction with the light wave (1). Summation over repeated pairs of matrix indices (FM_F) is implied.

The initial time in the solution of Eqs. (4)–(6) is

$$t'_0=t_0+\mathbf{k}(\mathbf{r}-\mathbf{r}_0)/\omega \tag{7}$$

at which time the leading edge of the light wave (1) arrives at the observation point $\mathbf{r}$, taking into account the delay of the light signal in the volume containing the atomic gas under study. In this regard, it is logical to put $t'=t-t'_0$ in the argument of the amplitude $a(t')$ of the field $\mathbf{E}$ in Eqs. (4)–(6), as well as in the density matrix $\rho=\rho(t')$. In the volume considered, prior to the initial time (7), the gas at an arbitrary observation point $\mathbf{r}$ is in statistical equilibrium, and the state of the atoms in a unit volume at this point can be described by the density matrix $\rho(0)$ in the FM_F representation,

$$\rho_{F_bM_{F_b},F_aM_{F_a}}(0)=\rho_{F_bM_{F_b},F'_bM'_{F'_b}}(0)=0,$$

$$\rho_{F_aM_{F_a},F'_aM'_{F'_a}}(0)=\frac{Nf(v)}{(2J_a+1)(2I+1)}\delta_{F_aF'_a}\cdot\delta_{M_{F_a}M'_{F_a}}, \tag{8}$$

where it is assumed that the hyperfine components of the ground state E_a are equally populated prior to the passage of the light wave, given that

$$\hbar|\Delta_{F_a}|\ll k_BT.$$

Equations (8) determine the normalization of the density matrix in Eqs. (4)– (6), according to which $\rho=\rho(t')$ describes the number of atoms per unit volume moving with velocity $\mathbf{v}$.

In contrast to atoms with zero nuclear spin,[9] the interaction of the light wave (1) with an atom in the presence of nuclear spin leads to a redistribution over the Zeeman sublevels not only in the resonance components $E_{F_a^0}$ and $E_{F_b^0}$ with total angular momenta F_a^0 and F_b^0, for which $E_{F_b^0}-E_{F_a^0}\approx\hbar\omega$, but also in a number of other hyperfine components E_{F_a} of the ground state E_a. This is due to spontaneous transitions from an excited state of the atom to lower states, which conform to the total angular momentum selection rules. Moreover, for weak splitting of the excited state E_b, the light wave (1) can interact with several hyperfine components E_{F_b} of the excited state E_b. Thus, in the general case all hyperfine components of the excited state E_b and the ground state E_a contribute to the photoinduced magnetization, and the total photoinduced magnetization is

$$\boldsymbol{\mu}=\boldsymbol{\mu}_b+\boldsymbol{\mu}_a, \tag{9}$$

where μ_b and μ_a are the photoinduced magnetizations of E_b and E_a are described by the well-known formulas

$$\boldsymbol{\mu}_b=-\mu_B\int g_{F_b}\mathbf{F}_{F_bM_{F_b},F_bM'_{F_b}}\rho_{F_bM'_{F_b},F_bM_{F_b}}d\mathbf{v}, \tag{10}$$

$$\boldsymbol{\mu}_a=-\mu_B\int g_{F_a}\mathbf{F}_{F_aM_{F_a},F_aM'_{F_a}}\rho_{F_aM'_{F_a},F_aM_{F_a}}d\mathbf{v}, \tag{11}$$

where

$$\mu_B=|e|\hbar/2mc,$$

$$g_{F_b}=\frac{F_b(F_b+1)+J_b(J_b+1)-I(I+1)}{2F_b(F_b+1)}\times g_{J_b}-\frac{F_b(F_b+1)+I(I+1)-J_b(J_b+1)}{2F_b(F_b+1)}g_I, \tag{12}$$

$F_{F_bM_{F_b},F_bM'_{F_b}}$ is the matrix element of the total angular momentum operator $\mathbf{F}$, μ_B is the Bohr magneton, e and m are the electron charge and mass, g_{J_b} is the Landé factor or the g factor of the electronic subsystem, and g_I is the nuclear g factor. The physical quantities in Eq. (11) have the same meaning, and differ only in the replacement $b\to a$. Specifically, the most important quantity g_{F_a} is given by Eq. (12) with $b\to a$ in all physical quantities.

To calculate Eq. (9), the solution of Eqs. (4)–(6) for the density matrix $\rho(t')$ in the weak field (1), according to perturbation theory, was represented by the finite series

$$\rho(t')=\rho(0)+\rho^{(1)}(t')+\rho^{(2)}(t'), \tag{13}$$

where the density matrix $\rho(0)$ is defined by (8), and $\rho^{(1)}(t')$ and $\rho^{(2)}(t')$ are linear and quadratic functions of the field **E**. Since $\rho(0)$ and $\rho^{(1)}(t')$ do not contribute to $\boldsymbol{\mu}$, the problem reduces to calculating $\rho^{(2)}(t')$.

In perturbation theory, the accuracy with which the sum (13) can be calculated, and the domain of applicability of the result, are different for the nonstationary and stationary regimes. For example, we can set $\gamma_a=0$ for the ground state of an atom in a highly tenuous gas. Then for the extended light wave (1) in the nonstationary state with $\gamma_a=0$, perturbation theory is applicable over the bounded time interval

$$\frac{|a(t')d_{F_bF_a}|^2\gamma_{ba}t'}{\hbar^2[(\omega-\omega_{F_bF_a}-ku)^2+\gamma_{ba}^2]}\ll 1, \tag{14}$$

where $d_{F_bF_a}$ is the reduced dipole moment.[13]

In the nonstationary regime with an ultrashort pulse (1) with duration τ for which

$$\tau\gamma_{ba}\ll 1, \tag{15}$$

the criterion for the applicability of perturbation theory at $0\leqslant t'$ in a gas of arbitrary density, irrespective of the numerical value of γ_a, is that this pulse have small area,[14]

$$\left|\hbar^{-2}d_{F_bF_a}\int_0^\tau a(t')dt'\right|^2\ll 1. \tag{16}$$

In a dense enough gas, where γ_a and γ_b are of the same order of magnitude, the perturbation expansion parameter for the steady state with constant amplitude a_0 is the left-hand side of the inequality

$$\frac{|a_0d_{F_bF_a}|^2\gamma_{ba}}{\hbar^2\gamma_a[(\omega-\omega_{F_bF_a}-ku)^2+\gamma_{ba}^2]}\ll 1, \tag{17}$$

which shows that the steady state is not attained when $\gamma_a=0$.

If γ_a and γ_b are of the same order of magnitude, then (17) will still hold in the nonstationary regime, with slowly varying amplitude $a(t')$ after the substitution $a_0\to a(t')$ with $0\leqslant t'$. We also note that to simplify the mathematics, an unimportant factor that does not exceed unity and depends on F_a and F_b was omitted from the left-hand sides of (14), (16), and (17).

3. MAGNETIZATION OF ATOMS IN AN EXCITED STATE

In the resonance approximation to the FM_F representation, the second term in Eq. (13) is required to be of the form

$$\rho^{(1)}_{F_bM_{F_b},F_aM_{F_a}}=r^{(1)}_{F_bM_{F_b},F_aM_{F_a}}\exp[i(\mathbf{kr}-\omega t)],$$

where the term omitted is, to order of magnitude, γ_{ba}/ω times less than the term retained. Moreover, the preexponential factor, like the third term in Eq. (13), is slowly varying compared with $\exp[i(\mathbf{k}\cdot\mathbf{r}-\omega t)]$. This makes it possible to bring Eqs. (4)–(6) to a form suitable for slowly varying functions, which in turn makes it possible to apply the method of successive approximations:

$$\left(\frac{d}{dt}-i\Delta_{F_bF_a}+\gamma_{ba}\right)r^{(1)}_{F_bM_{F_b},F_aM_{F_a}}=\frac{iNf(v)a(t')}{\hbar(2J_a+1)(2I+1)}\mathbf{l}_{\mathbf{k}\lambda}\mathbf{d}_{F_bM_{F_b},F_aM_{F_a}}, \tag{18}$$

$$\left(\frac{d}{dt}+i\omega_{F_bF_b'}+\gamma_b\right)\rho^{(2)}_{F_bM_{F_b},F_b'M_{F_b'}'}=\frac{i}{\hbar}[a(t')\mathbf{l}_{\mathbf{k}\lambda}\mathbf{d}_{F_bM_{F_b},F_aM_{F_a}}r^{(1)}_{F_aM_{F_a},F_b'M_{F_b'}'}(t')-a^*(t')r^{(1)}_{F_bM_{F_b},F_aM_{F_a}}(t')\mathbf{l}^*_{\mathbf{k}\lambda}\mathbf{d}_{F_aM_{F_a},F_b'M_{F_b'}'}], \tag{19}$$

$$\left(\frac{d}{dt}+i\omega_{F_aF_a'}+\gamma_a\right)\rho^{(2)}_{F_aM_{F_a},F_a'M_{F_a'}'}=\frac{i}{\hbar}[a^*(t')\mathbf{l}^*_{\mathbf{k}\lambda}\mathbf{d}_{F_aM_{F_a},F_bM_{F_b}}r'_{F_bM_{F_b},F_a'M_{F_a'}'}-a(t')r^{(1)}_{F_aM_{F_a},F_bM_{F_b}}\mathbf{l}_{\mathbf{k}\lambda}\mathbf{d}_{F_bM_{F_b},F_a'M_{F_a'}'}]+\frac{\gamma(2J_b+1)}{|d_{ba}|^2}\times\mathbf{d}_{F_aM_{F_a},F_bM_{F_b}}\rho^{(2)}_{F_bM_{F_b},F_b'M_{F_b'}}\mathbf{d}_{F_b'M_{F_b'},F_a'M_{F_a'}'}, \tag{20}$$

where

$$\Delta_{F_bF_a}=\omega-\omega_{F_bF_a}-\mathbf{kv},\quad \omega_{F_bF_b'}=\Delta_{F_b}-\Delta_{F_b'},$$

$$\omega_{F_aF_a'}=\Delta_{F_a}-\Delta_{F_a'}.$$

In Eqs. (18)–(20) we have for the slowly varying functions $r^{(1)}(t')$ and $\rho^{(2)}(t')$

$$\left(\frac{\partial}{\partial t}+\mathbf{v}\nabla\right)r^{(1)}(t')=\left(1-\frac{\mathbf{vk}(t')}{\omega}\right)\frac{dr^{(1)}(t')}{dt'}=\frac{dr^{(1)}(t')}{dt'},$$

$$\left(\frac{\partial}{\partial t}+\mathbf{v}\nabla\right)\rho^{(2)}(t')=\frac{d}{dt'}\rho^{(2)}(t'),$$

where matrix indices have been omitted for clarity, and terms of order v/c have likewise been neglected. Moreover, at the initial time $t'=0$, the desired functions in Eqs. (18)–(20) are equal to zero.

The photoinduced magnetization can be calculated most simply at the excited level E_b. Substituting the solution of Eq. (18)

$$r^{(1)}_{F_bM_{F_b},F_aM_{F_a}}(t')=\left[\frac{iNf(v)}{\hbar(2J_a+1)(2I+1)}\right]\times\exp[-(\gamma_{ba}-i\Delta_{F_bF_a})t']\times\int_0^{t'}d\tau_1\exp[(\gamma_{ba}-i\Delta_{F_bF_a})\tau_1]\times a(\tau_1)\mathbf{l}_{\mathbf{k}\lambda}\mathbf{d}_{F_bM_{F_b},F_aM_{F_a}}$$

into Eq. (19), the density matrix can easily be calculated in second-order perturbation theory, which makes it possible with the help of Eq. (10) to write

$$\boldsymbol{\mu}_b(t')=-\frac{\mu_B N I_b(t')}{\hbar^2(2J_a+1)(2I+1)}\, g_{F_b}\mathbf{F}_{F_bM_{F_b},F_bM'_{F_b}} \times(\mathbf{l}_{\mathbf{k}\lambda}\mathbf{d}_{F_bM_{F'_b},F_aM_{F_a}})(\mathbf{l}^*_{\mathbf{k}\lambda}\mathbf{d}_{F_aM_{Fa},F_bM_{F_b}}), \tag{21}$$

where

$$I_b(t')=\exp(-\gamma_b t')\int d\mathbf{v}f(v)\Bigg\{\int_0^{t'}d\tau_2 a^*(\tau_2) \times\exp[(\gamma_b-\gamma_{ba}+i\Delta_{F_bF_a})\tau_2]\int_0^{\tau_2}d\tau_1 a(\tau_1) \times\exp[(\gamma_{ba}-i\Delta_{F_bF_a})\tau_1]+\int_0^{t'}d\tau_2 a(\tau_2) \times\exp[(\gamma_b-\gamma_{ba}-i\Delta_{F_bF_a})\tau_2]\int_0^{\tau_2}d\tau_1 a^*(\tau_1) \times\exp[(\gamma_{ba}+i\Delta_{F_bF_a})\tau_1]\Bigg\}. \tag{22}$$

The summation in Eq. (21) over the projections of the total angular momenta is performed by going to $3j$ symbols $\left(\begin{smallmatrix} a & b & c \\ d & e & h \end{smallmatrix}\right)$ via the Wigner–Eckart theorem,[13] and using the contraction rules for $3j$ symbols. The result is

$$\mathbf{F}_{F_bM_{F_b},F_bM'_{F_b}}(\mathbf{l}_{\mathbf{k}\lambda}\mathbf{d}_{F_bM'_{F_b},F_aM_{F_a}})(\mathbf{l}^*_{\mathbf{k}\lambda}\mathbf{d}_{F_aM_{F_a},F_bM_{F_b}}) =\frac{i}{\sqrt{6}}[\mathbf{l}_{\mathbf{k}\lambda}\mathbf{l}^*_{\mathbf{k}\lambda}](-1)^{2F_b}F_{F_bF_b}d_{F_bF_a}d_{F_aF_b} \times\begin{Bmatrix} 1 & 1 & 1 \\ F_b & F_b & F_a \end{Bmatrix},$$

where the $6j$ symbol $\left\{\begin{smallmatrix} a & b & c \\ d & e & h \end{smallmatrix}\right\}$ is defined in Ref. 13, and the reduced angular momenta take the form

$$F_{F_bF_b}=\sqrt{F_b(F_b+1)(2F_b+1)},$$

$$d_{F_aF_b}=(-1)^{F_b-F_a}d^*_{F_bF_a},$$

$$d_{F_bF_a}=(-1)^{J_b+F_a+I+1}\sqrt{2F_b+1} \times\sqrt{2F_a+1}\begin{Bmatrix} J_b & F_b & I \\ F_a & J_a & 1 \end{Bmatrix}d_{ba}.$$

Finally, the photoinduced magnetization (10) on the excited level E_b can be written

$$\boldsymbol{\mu}_b(t')=-\frac{\mathbf{k}}{k}\lambda\beta\sin(2\psi)\,\frac{\mu_B N}{6\hbar^2(2J_a+1)(2I+1)} \times\sum_{F_bF_a}|d_{F_bF_a}|^2 g_{F_b}Q_b(F_b,F_a)I_b(t'), \tag{23}$$

where the summation over the total angular momenta is displayed explicitly and

$$Q_b(F_b,F_a)=\sqrt{6}(-1)^{F_a+F_b}\sqrt{F_b(F_b+1)(2F_b+1)} \times\begin{Bmatrix} 1 & 1 & 1 \\ F_b & F_b & F_a \end{Bmatrix}. \tag{24}$$

4. MAGNETIZATION OF ATOMS IN THE GROUND STATE

The calculation of the photoinduced magnetization of atoms in the ground- state energy level E_a is more difficult because atoms arrive in the sublevels of this state after spontaneous decay of the excited state of the atom. In contrast to Eq. (19) above, the solution of Eq. (20) contains two fundamentally different terms:

$$\rho^{(2)}_{F_aM_{F_a},F'_aM'_{F'_a}}(t')=\rho^{\mathrm{I}}_{F_aM_{F_a},F'_aM'_{F'_a}}(t')+\rho^{\mathrm{II}}_{F_aM_{F_a},F'_aM'_{F'_a}}(t'), \tag{25}$$

where the first is determined by the first term in brackets on the right-hand side of Eq. (20). Likewise, the second term in Eq. (25) depends on the second term on the right-hand side of Eq. (20), which describes the arrival of atoms in the ground state E_a as a result of the spontaneous emission of a photon in the excited state of the atom. The photoinduced magnetization (11) in the ground state E_a therefore also separates into two terms,

$$\boldsymbol{\mu}_a(t')=\boldsymbol{\mu}^{\mathrm{I}}_a(t')+\boldsymbol{\mu}^{\mathrm{II}}_a(t'), \tag{26}$$

which correspond to the first and second terms in Eq. (25).

The calculation of $\boldsymbol{\mu}^{\mathrm{I}}_a(t')$ is similar to that of $\boldsymbol{\mu}_b(t')$. The vector $\boldsymbol{\mu}^{\mathrm{I}}_a(t')$ is therefore equal to the expression (23) with the replacements $b\to a$ and $a\to b$ in all physical quantities in the double sum over F_b and F_a.

To calculate the second term in Eq. (26)

$$\boldsymbol{\mu}^{\mathrm{II}}_a(t')=-\mu_B\int g_{F'_a}\mathbf{F}_{F'_aM_{F'_a},F'_aM'_{F'_a}}\rho^{\mathrm{II}}_{F'_aM'_{F'_a},F'_aM_{F'_a}}(t')d\mathbf{v}, \tag{27}$$

it is necessary to solve Eq. (20), taking into account only the second term with the factor γ on the right-hand side, and to substitute the resulting solution $\rho^{\mathrm{II}}_{F'_aM'_{F'_a},F'_aM_{F'_a}}(t')$ obtained into Eq. (27), yielding

$$\boldsymbol{\mu}^{\mathrm{II}}_a(t')=-\frac{\mu_B\gamma(2J_b+1)N}{\hbar^2(2J_a+1)(2I+1)|d_{ba}|^2} \times g_{F'_a}\mathbf{F}_{F'_aM_{F'_a},F'_aM'_{F'_a}}\exp(-\gamma_a t')\int_0^{t'}d\tau_3 \times\exp(\gamma_a\tau_3)(\mathbf{d}_{F'_aM'_{F'_a},F_bM_{F_b}}\rho^{(2)}_{F_bM_{F_b},F'_bM_{F'_b}}(\tau_3) \times\mathbf{d}_{F'_bM_{F'_b},F'_aM_{F'_a}}). \tag{28}$$

If the solution of Eq. (19) is used for $\rho^{(2)}_{F_bM_{F_b},F'_bM_{F'_b}} \times(\tau_3)$, then Eq. (28) assumes the complicated form

$$\boldsymbol{\mu}_a^{\mathrm{II}}(t')=-\frac{\mu_B\gamma(2J_b+1)N\mathbf{S}}{\hbar^2(2J_a+1)(2I+1)|d_{ba}|^2}$$
$$\times\exp(-\gamma_a t')\int_0^{t'}d\tau_3$$
$$\times\exp[(\gamma_a-\gamma_b-i\omega_{F_bF_b'})\tau_3]\int_0^{\tau_3}d\tau_2F(\tau_2)$$
$$\times\exp[(\gamma_b+i\omega_{F_bF_b'})\tau_2], \qquad (29)$$

where

$$\mathbf{S}=g_{F_a'}\mathbf{F}_{F_a'M_{F_a'},F_a'M_{F_b'}'}(\mathbf{d}_{F_a'M_{F_a'}',F_bM_{F_b}}(\mathbf{l}_{\mathbf{k}\lambda}\mathbf{d}_{F_bM_{F_b},F_aM_{F_a}})$$
$$\times(\mathbf{l}_{\mathbf{k}\lambda}^*\mathbf{d}_{F_aM_{F_a}F_b'M_{F_b'}})\mathbf{d}_{F_b'M_{F_b'},F_a'M_{F_a'}'}, \qquad (30)$$

$$F(\tau_2)=\int d\mathbf{v}f(v)\Big\{a^*(\tau_2)\exp[-(\gamma_{ba}-i\Delta_{F_bF_a})\tau_2]$$
$$\times\int_0^{\tau_2}d\tau_1a(\tau_1)\exp[(\gamma_{ba}-i\Delta_{F_bF_a})\tau_1]$$
$$+a(\tau_2)\exp[-(\gamma_{ba}+i\Delta_{F_b'F_a})\tau_2]$$
$$\times\int_0^{\tau_2}d\tau_1a^*(\tau_1)\exp[(\gamma_{ba}+i\Delta_{F_b'F_a})\tau_1]\Big\}. \qquad (31)$$

In Eq. (30) it is necessary to sum over the projections of the total angular momenta in using the Wigner–Eckart theorem and the contraction rules for the $3j$ symbols. This brings the vector $\mathbf{S}$ to the form

$$\mathbf{S}=\frac{i}{\sqrt{6}}[\mathbf{l}_{\mathbf{k}\lambda}\mathbf{l}_{\mathbf{k}\lambda}^*]g_{F_a'}F_{F_a'F_a'}d_{F_a'F_b}d_{F_bF_a}d_{F_aF_b'}d_{F_b'F_a'}$$
$$\times\begin{Bmatrix}1 & 1 & 1\\ F_b & F_b' & F_a\end{Bmatrix}\begin{Bmatrix}F_a' & 1 & F_a'\\ F_b & 1 & F_b'\end{Bmatrix}.$$

Moreover, to simplify Eq. (29) it is necessary to integrate over τ_3. As a result, the vector $\boldsymbol{\mu}_a^{II}(t')$ can be written in the form

$$\boldsymbol{\mu}_a^{II}(t')=-(\mathbf{k}/k)\lambda\beta\sin(2\psi)\frac{\mu_BN|d_{ba}|^2}{6\hbar^2(2J_a+1)(2I+1)}$$
$$\times\sum_{F_aF_bF_a'F_b'}g_{F_a'}Q(F_a,F_a',F_b,F_b')I_{F_bF_b'}(t'), \qquad (32)$$

where the summation over the total angular momenta is displayed explicitly, and we have introduced the quantities

$$Q(F_a,F_a',F_b,F_b')=\frac{\sqrt{6}}{|d_{ba}|^4}F_{F_a'F_a'}d_{F_a'F_b}d_{F_bF_a}d_{F_aF_b'}d_{F_b'F_a'}$$
$$\times(2J_b+1)\begin{Bmatrix}1 & 1 & 1\\ F_b & F_b' & F_a\end{Bmatrix}$$
$$\times\begin{Bmatrix}F_a' & 1 & F_a'\\ F_b & 1 & F_b'\end{Bmatrix}, \qquad (33)$$

$$I_{F_bF_b'}(t')=\frac{\gamma}{\gamma_a-\gamma_b-i\omega_{F_bF_b'}}\Big\{\exp[-(\gamma_b+i\omega_{F_bF_b'})t']$$
$$\times\int_0^{t'}d\tau_2F(\tau_2)\exp[(\gamma_b+i\omega_{F_bF_b'})\tau_2]$$
$$-\exp(-\gamma_at')\int_0^{t'}d\tau_2F(\tau_2)\exp(\gamma_a\tau_2)\Big\}. \qquad (34)$$

The two terms determined in Eq. (26) solve the problem of the photomagnetization of an atom in the ground state, with energy E_a.

5. GENERAL FORMULA FOR PHOTOINDUCED MAGNETIZATION

Using the results obtained for the photoinduced magnetizations (23), (26), and (32) in the excited state E_b and the ground state E_a, we finally find the total photoinduced magnetization (9) of an atomic gas, which describes both the nonstationary and stationary regimes:

$$\boldsymbol{\mu}(t')=-\frac{\mathbf{k}}{k}\lambda\beta\sin(2\psi)\frac{\mu_bN|d_{ba}|^2}{6\hbar^2(2J_a+1)(2I+1)}$$
$$\times\sum_{F_bF_a}\Big\{\left|\frac{d_{F_bF_a}}{d_{ba}}\right|^2[g_{F_a}Q_a(F_a,F_b)I_a(t')$$
$$+g_{F_b}Q_b(F_b,F_a)I_b(t')]$$
$$+\sum_{F_a'F_b'}g_{F_a'}Q(F_a,F_a',F_b,F_b')I_{F_bF_b'}(t')\Big\}, \qquad (35)$$

where $Q_a(F_a,F_b)$ and $I_a(t')$ are obtained from Eqs. (24) and (22) by interchanging the indices b and a in all physical quantities, and using the fact that $\gamma_{ab}=\gamma_{ba}$.

In the absence of nuclear spin, with $I=\omega_{F_bF_b'}=0$, $E_{F_a}=E_a$, $E_{F_b}=E_b$, $F_a=J_a$, and $F_b=J_b$, the general Eq. (35) transforms into the corresponding formula[9] for a gas of atoms with zero nuclear spin, if in the latter we set $N_a=N$ and $N_b=0$. In the presence of nuclear spin, the magnetic moment (35) is nonzero only for a light wave (1) with a rotating electric field, i.e., for circular or elliptical polarization.

We emphasize that three terms of very different physical origin are obtained when the braces in Eq. (35) are expanded. The first and second, with the factors g_{F_a} and g_{F_b}, describe the magnetization of an atom's hyperfine components in the ground state E_a and excited state E_b when the atom interacts with the light wave (1). The third term, with the factor $g_{F_a'}$, describes the magnetization of the atom in the hyperfine sublevels of the ground state E_a due to the atom's arrival at these sublevels via spontaneous emission of a photon from an excited state belonging to the level E_b. This process occurs both during the passage of the light wave (1) and after it has passed, and it engenders the optical pumping effect in the photoinduced magnetization.

The general Eq. (35) is suited to any separation between hyperfine components, and arbitrary Doppler widths ku and

relaxation constants γ, γ_a, and γ_b. However, it can be simplified in some cases of practical import. For show this, we employ the fact that the quantity given by (33) is real and is invariant under $F_b \to F_b'$ and $F_b' \to F_b$. The integral (34) transforms under $F_b \to F_b'$ and $F_b' \to F_b$ into the complex-conjugate expression. Consequently, in the double sum over F_b and F_b' in Eq. (35), because of the factor (33), only the real part of the integral $I_{F_bF_b'}(t')$, which is invariant under $F_b \to F_b'$ and $F_b' \to F_b$, remains. This makes it possible to simplify the expression (35) when

$$\omega_{F_bF_b'}^2 \geqslant (\Delta_{F_b+1}-\Delta_{F_b})^2 \gg \gamma^2, \tag{36}$$

By virtue of the factor preceding the braces in (34), when $\gamma_b \sim \gamma_a \sim \gamma$, the real part of the integral $I_{F_bF_b'}(t')$, which is even under $F_b \to F_b'$ and $F_b' \to F_b$, can be written as

$$I_{F_bF_b'}(t') = \frac{\gamma}{\gamma_b-\gamma_a}[I_a(t')-I_b(t')]\delta_{F_bF_b'}, \tag{37}$$

where the terms omitted are a factor $\gamma^2/\omega_{F_bF_b'}^2$ down from those remaining in Eq. (37). If the stronger inequality

$$\omega_{F_bF_b'}^2 \geqslant (\Delta_{F_b+1}-\Delta_{F_b})^2 \gg \gamma_b^2, \gamma_{ba}^2 \tag{38}$$

holds, with $\gamma_b > \gamma$, then the real part of the integral $I_{F_bF_b'}(t')$, which is even under $F_b \to F_b'$ and $F_b' \to F_b$, can be expanded in powers of the small parameters $\gamma_b/\omega_{F_bF_b'}$ and $\gamma_{ba}/\omega_{F_bF_b'}$, which will appear in this series in various combinations with even powers of $\omega_{F_bF_b'}$. Finally, we arrive at Eq. (37), in which the omitted terms are negligible, since they are of the same order as $\gamma_b^2/\omega_{F_bF_b'}^2$, $\gamma_b\gamma_{ba}/\omega_{F_bF_b'}^2$, and $\gamma_{ba}^2/\omega_{F_bF_b'}^2$.

Substituting Eq. (37), the general formula (35) simplifies appreciably and assumes a form convenient for practical calculations:

$$\begin{aligned}\boldsymbol{\mu}(t') = -\frac{\mathbf{k}}{k}\lambda\beta\sin(2\psi)\frac{\mu_B N}{6\hbar^2(2J_a+1)(2I+1)} \\ \times\sum_{F_aF_b}|d_{F_bF_a}|^2\Big\{g_{F_a}Q_a(F_a,F_b)I_a(t') \\ +g_{F_b}Q_b(F_b,F_a)I_b(t')+\frac{\gamma}{\gamma_b-\gamma_a}[I_a(t') \\ -I_b(t')]\sum_{F_a'}g_{F_a'}Q(F_a,F_a',F_b)\Big\},\end{aligned} \tag{39}$$

where

$$Q(F_a,F_a',F_b) = |d_{F_bF_a}|^2|d_{ba}|^{-2}Q(F_a,F_a',F_b),$$

$$\begin{aligned}Q(F_a,F_a',F_b) = \sqrt{6}\,(-1)^{F_a-F_a'}(2J_b+1) \\ \times\sqrt{F_a'(F_a'+1)(2F_a'+1)}(2F_a'+1) \\ \times(2F_b+1) \\ \times\begin{Bmatrix}J_b & F_b & I\\ F_a' & J_a & 1\end{Bmatrix}^2\begin{Bmatrix}1 & 1 & 1\\ F_b & F_b & F_a\end{Bmatrix} \\ \times\begin{Bmatrix}F_a' & 1 & F_a'\\ F_b & 1 & F_b\end{Bmatrix}.\end{aligned} \tag{40}$$

Because of the inequalities (36) and (38), Eq. (39) has no analogs among the photoinduced magnetizations obtained in Refs. 9 and 10 for a gas of atoms with zero nuclear spin. In this vein, we call attention to certain interesting special cases due solely to hyperfine splitting. If the hyperfine splitting in the ground state E_a and excited state E_b is large enough,

$$|\Delta_{F_a+1}-\Delta_{F_a}|, |\Delta_{F_a+1}-\Delta_{F_b}| \gg \gamma_{ba}, \ ku, \tag{41}$$

the frequency ω of the light wave (1) can be chosen such that only two sublevels $E_{F_b^0}$ and $E_{F_a^0}$ of the hyperfine structures under study are in resonance:

$$\omega = (F_{F_b^0}-E_{F_a^0})\hbar^{-1}. \tag{42}$$

Then, on account of the integrals $I_a(t')$ and $I_b(t')$ in the simplified Eq. (39), the double sum over F_a and F_b vanishes and only the resonance term with $F_a = F_a^0$ and $F_b = F_b^0$ remains. In this case, the term with the factor γ in Eq. (39) takes into account, by virtue of the sum over F_a', the magnetization of the atom not only in the resonant sublevel $E_{F_a^0}$, but also in other nonresonant sublevels $E_{F_a'}$ of the ground state E_a ($F_a' \neq F_a^0$) in which the atoms appeared as a result of optical pumping by the light wave (1).

Thus, in the presence of nuclear spin, the term in Eq. (6) with the factor γ incorporates the optical pumping effect in the photoinduced magnetization (39). This effect increases or decreases the total photoinduced magnetization (39), depending on the angular momenta and the number of hyperfine components in the ground state E_a and excited state E_b. If the frequency ω is swept while the inequalities (41) hold, then other pairs of sublevels E_{F_b} and E_{F_a} allowed by the selection rules for the total angular momentum, and in which the optical pumping effect is prominent, will also be in resonance at the frequency given by (42). An example in which the inequalities (41) hold are the $6^2S_{1/2}$ and $6^2P_{1/2}^0$ terms of $^{133}_{55}$Cs with spin $I = 7/2$.

The general equation (35) also contains the optical pumping effect in the photoinduced magnetization in the term with the factor $g_{F_a'}$. However, this effect is most pronounced in the case of the simplified formula (39).

The general equation (35) can also be simplified for the transition $J_a \to J_b = J_a + 1$ and $J_a \geqslant I$ for large hyperfine splittings only in the ground state,

$$|\Delta_{F_a+1} - \Delta_{F_a}| \gg \gamma_{ba}, ku, \tag{43}$$

whereupon the sublevel $E_{F_a^0}$ with the lowest total angular momentum $F_a^0 = J_a - I$ is in resonance (42). In this case, by virtue of the total angular momentum selection rule, only the hyperfine component with the lowest total angular momentum $F_b^0 = J_a - I + 1$, with arbitrary splitting between the hyperfine components and irrespective of the inequalities (36) and (38), can be in the excited state E_b. We must then put $F_a = F_a^0$ and $F_b = F_b^0$ in the general equation (35), bringing it to the simplified form (39) with $F_a = F_a^0$ and $F_b = F_b^0$. The optical pumping effect in the photoinduced magnetization remains in force. An example is provided by the $4^8S_{7/2}^0$ and $6^8P_{9/2}$ terms of $^{152}_{63}$Eu with spin $I = 5/2$.

A similar situation arises in the $J_a = J_b + 1 \to J_b$ transition for arbitrary nuclear spin I when the inequalities (43) hold, if the sublevel $E_{F_a^0}$ with the maximum total angular momentum $F_a^0 = J_a + I = J_b + I + 1$ is in resonance (42). By virtue of the total angular momentum selection rule, the light wave (1) excites in the upper state E_b only one hyperfine component $E_{F_b^0}$ with the maximum total angular momentum $F_b^0 = J_b + I$, regardless of the inequalities (36) and (38). The general equation (35) then goes into (39) with $F_a = F_a^0$ and $F_b = F_b^0$, with the optical pumping effect remaining in the photoinduced magnetization. An example is provided by the $4^2D_{3/2}$ and $5^2P_{1/2}^0$ terms of $^{89}_{39}$Y with spin $I = 1/2$.

Finally, we consider a different limiting case from (38), in which the hyperfine splitting in the excited state E_b is small compared to the width $\hbar\gamma_b$ of the level,

$$\omega_{F_bF_b'}^2 \ll \gamma_b^2, \tag{44}$$

while conversely, in the ground state E_a we have

$$|\Delta_{F_a+1} - \Delta F_a| \gg \gamma_{ba}. \tag{45}$$

We must then put $\omega_{F_bF_b'} = 0$ in the integral (34) and use the fact that (33) is invariant under $F_b \to F_b'$ and $F_b' \to F_b$. The general equation (35) then becomes

$$\boldsymbol{\mu}(t') = -\frac{\mathbf{k}}{k}\lambda\beta \sin(2\psi)\frac{\mu_B N|d_{ba}|^2}{6\hbar^2(2J_a+1)(2I+1)} \times \sum_{F_aF_b}\left\{\left|\frac{d_{F_bF_a}}{d_{ba}}\right|^2 [g_{F_a}Q_a(F_a,F_b)I_a(t') + g_{F_b}Q_b(F_b,F_a)I_a(t')] + \frac{\gamma}{\gamma_b - \gamma_a}[I_a(t') - I_b(t')]\sum_{F_a'F_b'} g_{F_a'}Q(F_a,F_a',F_b,F_b')\right\}. \tag{46}$$

6. PHOTOINDUCED MAGNETIZATION IN THE STEADY STATE

Let the light wave (1) with the stepped profile

$$a(t') = 0 \quad \text{when } t' < 0,$$

$$a(t') = a_0 \exp(-i\alpha) \quad \text{when } t' \geqslant 0, \tag{47}$$

propagate in an atomic gas; here a_0 and α are constants. Then at the start of the nonstationary state in the brief time interval

$$\gamma_a t' \ll 1, \quad \gamma_b t' \ll 1, \quad |\omega_{F_bF_b'}|t' \ll 1,$$

$$|\omega - \omega_{F_bF_a}|t' \ll 1, \quad kut' \ll 1$$

the general equation (35) simplifies considerably, and the nonstationary photoinduced magnetization increases quadratically with time:

$$\boldsymbol{\mu}(t') = -\frac{1}{2}\frac{\mathbf{k}}{k}\lambda\beta \sin(2\psi)M_{ba}(\gamma_{ba}t')^2 \times \sum_{F_aF_b}(2F_a+1)(2F_b+1)\begin{Bmatrix} J_b & F_b & I \\ F_a & J_a & 1 \end{Bmatrix}^2 \times [g_{F_a}Q_a(F_a,F_b) + g_{F_b}Q_b(F_b,F_a)],$$

where M_{ba} is a constant with dimensions of magnetic moment per unit volume:

$$M_{ba} = \mu_B N|d_{ba}|^2 a_0^2/3\hbar^2\gamma_{ba}^2(2J_a+1)(2I+1).$$

For a highly tenuous gas of atoms with relaxation constants $\gamma_a \; ll \gamma_b$, when the inequalities (38) hold, there exists a long time interval

$$1/\gamma_b \ll t' \ll 1/\gamma_a \tag{48}$$

over which the nonstationary photoinduced magnetization (39) increases linearly with time,

$$\boldsymbol{\mu}(t') = -(\mathbf{k}/k)\lambda\beta \sin(2\psi)M_{ba}\gamma_{ba}t' \times \sum_{F_aF_b}(2F_a+1)(2F_b+1)\begin{Bmatrix} J_b & F_b & I \\ F_a & J_a & 1 \end{Bmatrix}^2 \times \left[g_{F_a}Q_a(F_a,F_b) + \frac{\gamma}{\gamma_b}\sum_{F_\alpha'} g_{F_a'}Q(F_a,F_a',F_b)\right] I(\Delta_{F_bF_a}), \tag{49}$$

where

$$I(\Delta_{F_bF_a}) = \int \frac{f(v)\gamma_{ba}^2 d\mathbf{v}}{\Delta_{F_bF_a}^2 + \gamma_{ba}^2}. \tag{50}$$

In particular, for a very tenuous gas we have $\gamma_a = 0$ and $\gamma_b = \gamma$, so Eq. (49) is bounded in time t' by (14) with $a(t') = a_0$ instead of the preceding inequality (48). For a weak light wave (1) with amplitude (47) this time interval with $\gamma_a = 0$ and $\gamma_b = \gamma$ is fairly long

$$\frac{1}{\gamma_b} \ll t' \ll \frac{\hbar^2[(\omega-\omega_{F_bF_a}-ku)^2+\gamma_{ba}^2]}{\gamma_{ba}a_0^2|d_{F_bF_a}|^2}. \qquad (51)$$

This makes it possible to investigate optical pumping in photoinduced magnetization (49) by sweeping the frequency ω and investigating experimentally the derivative $d\mu(t')/dt'$ in the time intervals (48) or (51). To do so, the light wave (1), in accordance with the experiments of Refs. 6–8, should pass through a cell that contains the experimental gas, and that is positioned inside the receiver detection coil. The variable photoinduced magnetization generates an electric current in the windings of the receiver coil, producing a potential difference U (an emf) at the ends of the coil. The emf is proportional to the time derivative $d\mu(t')/dt'$. Thus, the properties of the photoinduced magnetization of an atomic gas can be studied by investigating experimentally the electomotive force in the receiver coil.

In a atomic gas with $\gamma_a \sim \gamma_b$, at long times

$$1/\gamma_a \ll t', \quad 1/\gamma_b \ll t' \qquad (52)$$

the photoinduced magnetization (35) becomes stationary and assumes the constant value

$$\mu = -\frac{\mathbf{k}}{k}\lambda\beta \sin(2\psi) M_{ba} \sum_{F_aF_b} \left\{ \left|\frac{d_{F_bF_a}}{d_{ba}}\right|^2 I(\Delta_{F_bF_a}) \times \left[\frac{\gamma_{ba}}{\gamma_a} g_{F_a} Q_a(F_a,F_b) + \frac{\gamma_{ba}}{\gamma_b} g_{F_b} Q_b(F_b,F_a)\right] + \frac{\gamma\gamma_{ba}}{\gamma_a\gamma_b} \sum_{F_a'F_b'} g_{F_a'} Q(F_a,F_a',F_b,F_b') \times I(\Delta_{F_bF_a},\Delta_{F_b'F_a},\omega_{F_bF_b'}) \right\}, \qquad (53)$$

where

$$I(\Delta_{F_bF_a},\Delta_{F_b'F_a},\omega_{F_bF_b'}) = \frac{\gamma_b\gamma_{ba}}{2}\int dv f(v) \frac{\gamma_b\gamma_{ba}(\Delta_{F_bF_a}^2+\Delta_{F_b'F_a}^2+2\gamma_{ba}^2)+\omega_{F_bF_b'}^2(\Delta_{F_bF_a}\Delta_{F_b'F_a}-\gamma_{ba}^2)}{(\Delta_{F_bF_a}^2+\gamma_{ba}^2)(\Delta_{F_b'F_a}^2+\gamma_{ba}^2)(\omega_{F_bF_b'}^2+\gamma_b^2)}. \qquad (54)$$

Note that Eq. (53) was also obtained by solving Eqs. (18)–(20) for the monochromatic wave (1) with constant amplitude a_0 in the steady state without using the initial conditions. The fact that the solution obtained in this manner is identical with the nonstationary solution (35) in the limiting case of long times (52) confirms the correctness of the result (53) found above.

When the hyperfine splitting in the excited state E_b satisfies the inequality (38), the integral (54) is identical with (50), if small terms of order $\gamma_b^2/\omega_{F_bF_b'}^2$, $\gamma_b\gamma_{ba}/\omega_{F_bF_b'}^2$, and $\gamma_{ba}^2/\omega_{F_bF_b'}^2$ with $F_b' \neq F_b$ are dropped. As a result, the stationary photoinduced magnetization (53) assumes a simplified practical form that can be conveniently used in the steady state:

$$\mu = -\frac{\mathbf{k}}{k}\lambda\beta \sin(2\psi) M_1 \sum_{F_aF_b} I(x,y)(2F_a+1)(2F_b+1) \times \begin{Bmatrix} J_b & F_b & I \\ F_a & J_a & 1 \end{Bmatrix}^2 \left[\frac{\gamma_{ba}}{\gamma_a}\tilde{g}_{F_a} Q_a(F_a,F_b) + \frac{\gamma_{ba} g_{J_b}}{\gamma_b g_{J_a}} \times \tilde{g}_{F_b} Q_b(F_b,F_a) + \frac{\gamma\gamma_{ba}}{\gamma_a\gamma_b}\sum_{F_a'} \tilde{g}_{F_a'} Q(F_a,F_a',F_b)\right], \qquad (55)$$

where

$$M_1 = \frac{g_{J_a}\gamma_{ba}^2 M_{ba}}{\sqrt{\pi}(ku)^2}, \quad I(x,y) = \int_{-\infty}^{\infty} \frac{\exp(-\xi^2)d\xi}{(x-\xi)^2+y^2},$$

$$\tilde{g}_{F_a} = \frac{F_a(F_a+1)+J_a(J_a+1)-I(I+1)}{2F_a(F_a+1)},$$

$$x = \frac{\omega-\omega_{F_bF_a}}{ku}, \quad y = \frac{\gamma_{ba}}{ku},$$

$\tilde{g}_{F_b}$ is obtained from $\tilde{g}_{F_a}$ by letting $a \to b$, with $g_I \ll g_{J_a}$, g_{J_b}. Depending on the type of transition $F_a \to F_b$, $Q_a(F_a,F_b)$ and $Q_b(F_b,F_a)$ assume the following values:

$F_a = F \to F_b = F (F>0)$: $Q_a(F_a,F_b) = Q_b(F_b,F_a) = 1$,

$F_a = F \to F_b = F+1$: $Q_a(F_a,F_b) = -F$,

$Q_b(F_b,F_a) = F+2$,

$F_a = F \to F_b = F-1 (F \geqslant 1)$: $Q_a(F_a,F_b) = F+1$,

$Q_b(F_b,F_a) = -F+1$.

On the other hand, for $F_a \to F_b$ and $J_a \to J_b$ with arbitrary total angular momenta and electronic angular momenta, $Q(F_a,F_a',F_b)$ does not simplify significantly, so Eq. (40) must be used in actual calculations of μ.

If the inequalities (44) and (45) hold, it is possible to make use of the relation

$$I(\Delta_{F_bF_a},\Delta_{F_b'F_a},0) = \tfrac{1}{2}[I(\Delta_{F_bF_a})+I(\Delta_{F_b'F_a})],$$

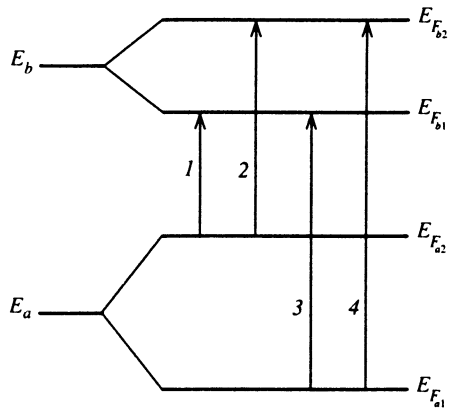

FIG. 1. Hyperfine structure for doublet splitting of the atomic ground state E_a and excited state E_b. The numbers *1*, *2*, *3*, and *4* label the allowed transitions between hyperfine components.

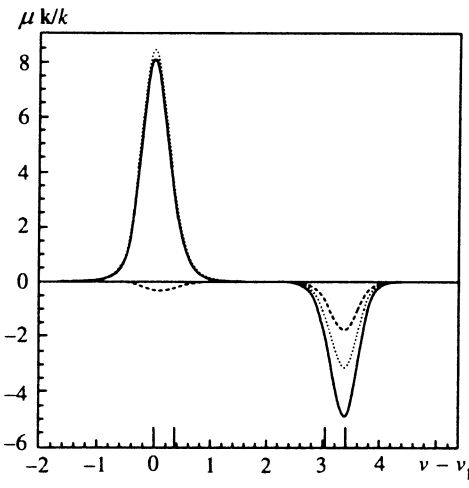

FIG. 2. Projection $\mu(t')\mathbf{k}/k$ of steady-state photoinduced magnetization (55) as a function of $\nu-\nu_1$. The horizontal unit is 1 GHz; the vertical unit is 50 $M_1/3$. The four long bars on the abscissa mark transition frequencies ν_1, ν_2, ν_3, and ν_4. Dotted curve: contribution of resonant sublevels neglecting optical pumping. Dashed curve: contribution of the sublevels populated by optical pumping (optical pumping effect). Solid curve: total contribution of all sublevels.

which transforms the general equation (53) for the steady-state photoinduced magnetization:

$$\mu=-\frac{\mathbf{k}}{k}\lambda\beta\sin(2\psi)M_1\sum_{F_aF_b}\left\{(2F_a+1)(2F_b+1)\right.$$
$$\times\begin{Bmatrix}J_b & F_b & I\\ F_a & J_a & 1\end{Bmatrix}^2\left[\frac{\gamma_{ba}}{\gamma_a}\tilde{g}_{F_a}Q_a(F_a,F_b)\right.$$
$$\left.+\frac{\gamma_{ba}g_{J_b}}{\gamma_b g_{J_a}}\tilde{g}_{F_b}Q_b(F_b,F_a)\right]$$
$$\left.+\frac{\gamma\gamma_{ba}}{\gamma_a\gamma_b}\sum_{F_a'F_b'}\tilde{g}_{F_a'}Q(F_a,F_a',F_b,F_b')\right\}I(x,y). \quad (56)$$

This equation can also be obtained from the nonstationary photoinduced magnetization (46) in the limiting case of long times (52).

The calculations in Eqs. (55) and (56) are simplest for doublet splitting, which is illustrated in Fig. 1. The optically allowed transitions, labeled 1, 2, 3, and 4, correspond to the following atomic transition frequencies:

$$\omega^{(1)}_{F_{b1}F_{a2}}=\omega_{ba}+\Delta_{F_{b1}}-\Delta_{F_{a2}}=2\pi\nu_1, \quad (57)$$

$$\omega^{(2)}_{F_{b2}F_{a2}}=\omega_{ba}+\Delta_{F_{b2}}-\Delta_{F_{a2}}$$
$$=\omega^{(1)}_{F_{b1}F_{a2}}+\Delta_{F_{b2}}-\Delta_{F_{b1}}=2\pi\nu_2, \quad (58)$$

$$\omega^{(3)}_{F_{b1}F_{a1}}=\omega_{ba}+\Delta_{F_{b1}}-\Delta_{F_{a1}}$$
$$=\omega^{(1)}_{F_{b1}F_{a2}}+\Delta_{F_{a2}}-\Delta_{F_{a1}}=2\pi\nu_3, \quad (59)$$

$$\omega^{(4)}_{F_{b2}F_{a1}}=\omega_{ba}+\Delta_{F_{b2}}-\Delta_{F_{a1}}$$
$$=\omega^{(1)}_{F_{b1}F_{a2}}+\Delta_{F_{a2}}-\Delta_{F_{a1}}+\Delta_{F_{b2}}-\Delta_{F_{b1}}=2\pi\nu_4, \quad (60)$$

where ν_1, ν_2, ν_3, and ν_4 are the transition frequencies in hertz.

For a light wave (1) with left-hand circular polarization we set

$$\lambda=-1, \quad \psi=\pi/4, \quad \beta=1. \quad (61)$$

We now apply Eq. (55) and Eqs. (57)–(61) to the $5^2S_{1/2}$ and $5^2P^0_{1/2}$ terms of $^{85}_{37}$Rb, with nuclear spin $I=5/2$, for which[15] $E_b-E_a=1.560$ eV and

$$(E_{F_{a2}}-E_{F_{a1}})h^{-1}=3036 \text{ MHz},$$
$$(E_{F_{b2}}-E_{F_{b1}})h^{-1}=362 \text{ MHz}, \quad (62)$$

where $F_{a1}=F_{b1}=2$ and $F_{a2}=F_{b2}=3$. The spontaneous emission coefficient is $\gamma=34\cdot10^6$ sec^{-1}. From (57)–(60), the transition frequency differences required for the calculations are

$$\nu_2-\nu_1=362 \text{ MHz}, \quad \nu_3-\nu_1=3036 \text{ MHz},$$
$$\nu_4-\nu_1=3398 \text{ MHz}. \quad (63)$$

The gas temperature is $T=300$ K. This temperature corresponds to a Doppler width $ku=1914\cdot10^6$ sec^{-1} for the light wave (1), which is tuned to the transition frequency ω_{ba}. In a tenuous gas, we set $\gamma_a=0.5\gamma$, $\gamma_b=1.5\gamma$, and $\gamma_{ba}=\gamma$.

Figure 2 shows the projection of the photoinduced magnetization (55) in the direction of $\mathbf{k}/k$ for various frequencies $\nu=\omega/2\pi$ of the left-hand circular monochromatic wave (1). In accordance with Eq. (3), because of the small hyperfine splitting in the excited state E_b, the photoinduced magnetizations merge into two separate bell-shaped peaks near the resonant frequencies $\nu=\nu_1$ and $\nu=\nu_2$ and near $\nu=\nu_3$ and $\nu=\nu_4$. This pairwise merging of photoinduced magnetizations is related to the Doppler broadening of the atomic transitions when

$$ku \gg \gamma_{ba}, \quad \nu_2 - \nu_1 \sim ku/2\pi, \quad \nu_4 - \nu_3 \sim ku/2\pi.$$

These two peaks are distinct by virtue of the fact that $\nu_4 - \nu_1 \gg ku/2\pi$, a result of the large hyperfine splitting in the ground state E_a. The photoinduced magnetizations in the sublevels populated via optical pumping near the transition frequencies ν_1 and ν_2 have the opposite sign from the contribution of the resonance sublevels, and thereby attenuate the first peak. In the second peak, the photoinduced magnetizations in all sublevels near the transition frequencies ν_3 and ν_4 have the same direction, opposite the photoinduced magnetization of the first peak. The differing directions of the photoinduced magnetizations that produce the first and second peaks are due to the differing total angular momenta in the sublevels that contribute to the magnetization of the atom. Since different sublevels of the hyperfine structure contribute to each peak, the appearance of such peaks when the frequency ν of the light wave (1) is swept reflects the existence of characteristic hyperfine structure of the photoinduced magnetization for atoms with nuclear spin I. If $I=0$, then when the frequency ν is swept through the transition frequency ω_{ba}, one bell-shaped peak of photoinduced magnetization is obtained instead of individual peaks.

7. MAGNETIZATION OF A GAS BY AN ULTRASHORT PULSE

The inequality (15) holds when the ultrashort pulse (1) with duration τ and center frequency ω propagates in a gas, so γ_b and γ_{ba} must be set zero in the integrand of the integral in Eq. (22). This makes it possible to represent the integral in the region $0 \leqslant t'$ in the form

$$I_b(t') = \exp(-\gamma_b t') \int d\mathbf{v} f(v) \left| \int_0^{t'} a(\xi) \times \exp(-i\Delta_{F_b F_a}\xi) d\xi \right|^2. \tag{64}$$

Moreover, γ_a and γ_{ba} must be set to zero in the integrand of $I_a(t')$, after which $I_a(t')$ assumes the form (64) with the substitution $\gamma_b \to \gamma_a$. For the same reason we also set $\gamma_{ba}=0$ in $F(\tau_2)$ of (31).

If the inequality (43) holds with γ_{ba} replaced by the spectral width Δ_ω of the ultrashort pulse, then previous comments regarding this inequality remain valid. However, below we consider atoms with large hyperfine splitting in the excited state,

$$\omega^2_{F_b F'_b} \geqslant (\Delta_{F_b+1} - \Delta_{F_b})^2 \gg \gamma_b^2, \Delta_\omega^2, \tag{65}$$

which makes it possible to use the integral (37). The simplified formula (39) for the photoinduced magnetization of the gas by an ultrashort pulse then assumes the simple form

$$\boldsymbol{\mu}(t') = -\frac{\mathbf{k}}{k} \lambda\beta \sin(2\psi) \sum_{F_a F_b} M(F_a, F_b, t') \times \left\{ g_{F_a} Q_a(F_a, F_b) \exp(-\gamma_a t') + g_{F_b} Q_b(F_b, F_a) \exp(-\gamma_b t') + \frac{\gamma}{\gamma_b - \gamma_a} \times [\exp(-\gamma_a t') - \exp(-\gamma_b t')] \times \sum_{F'_a} g_{F'_a} Q(F_a, F'_a, F_b) \right\}, \tag{66}$$

where

$$M(F_a, F_b, t') = \frac{\mu_B N |d_{F_b F_a}|^2}{6\hbar^2 (2J_a+1)(2I+1)} \int d\mathbf{v} f(v) \left| \int_0^{t'} a(\xi) \times \exp(-i\Delta_{F_b F_a}\xi) d\xi \right|^2. \tag{67}$$

To elucidate the properties of the photoinduced magnetization (66), we consider a Maxwell–Boltzmann velocity distribution $f(v)$ and Gaussian amplitude $a(t')$,

$$a(t') = a_0 \exp\left(-\frac{(t' - \tau/2)^2}{(2\tau_p)^2} - i\alpha \right), \tag{68}$$

where a_0 and α are real constants and τ_p is the characteristic time of the ultrashort pulse ($4\tau_p < \tau$). Here it is assumed that the amplitude $a(t')$ vanishes outside the time interval $0 \leqslant t' \leqslant \tau$, and the numerical value of τ is so chosen that the discarded parts of the Gaussian function in Eq. (68), which correspond to the time intervals $-\infty \leqslant t' \leqslant 0$ and $\tau \leqslant t' \leqslant \infty$, make a negligible contribution to the desired physical quantities when the amplitude (68) is used over the interval $0 \leqslant t' \leqslant \tau$. For example, for $\tau = 6\tau_p$, $\tau = 7\tau_p$, and $\tau = 8\tau_p$, the respective sums of integrals of the function (68) over the two deleted regions $-\infty \leqslant t' \leqslant 0$ and $\tau \leqslant t' \leqslant \infty$ are a factor of 0.035, 0.014, and 0.005 times less than the integral of the same function over the region $0 \leqslant t' \leqslant \tau$. For $\tau = 8\tau_p$, the spectral width Δ_ω of this ultrashort pulse with amplitude (68) is

$$\Delta_\omega = \sqrt{2 \ln 2}\, \tau_p^{-1}.$$

To investigate the photoinduced magnetization (66) at all times $0 \leqslant t'$, it is necessary to calculate the velocity integral in Eq. (66) using the amplitude (68). As a result, the photoinduced magnetization (66) acquires the following form at times $0 \leqslant t'$:

$$\mu(t')=-\frac{\mathbf{k}}{k}\lambda\beta\sin(2\psi)M_0\sum_{F_aF_b}I(t')(2F_a+1)$$
$$\times(2F_b+1)\begin{Bmatrix}J_b & F_b & I\\ F_a & J_a & 1\end{Bmatrix}^2\Bigg\{\tilde g_{F_a}Q_a(F_a,F_b)$$
$$\times\exp(-\gamma_a t')+\frac{g_{J_b}}{g_{J_a}}\tilde g_{F_b}Q_b(F_b,F_a)$$
$$\times\exp(-\gamma_b t')+\frac{\gamma}{\gamma_b-\gamma_a}[\exp(-\gamma_a t')$$
$$-\exp(-\gamma_b t')]\sum_{F_a'}\tilde g_{F_a'}Q(F_a,F_a',F_b)\Bigg\},\qquad(69)$$

where

$$M_0=(\gamma_{ba}\tau_p)^2 g_{J_a}M_{ba},$$
$$I(t')=\frac{1}{2\tau_p^2}\int_{-\pi/2}^{t'-\pi/2}d\xi\int_{-\pi/2}^{t'-\pi/2}d\eta$$
$$\times\cos[(\omega-\omega_{F_bF_a})(\xi-\eta)]$$
$$\times\exp\left(-\frac{\xi^2+\eta^2}{(2\tau_p)^2}-\frac{(\xi-\eta)^2}{(2\tau_D)^2}\right),\qquad(70)$$

and the time constant $\tau_D=1/ku$ is the reciprocal of the Doppler relaxation rate.

According to Eqs. (67) and (68), the integral (70) changes with time over the interval $0\leqslant t'\leqslant\tau$ and remains constant and equal to $I(\tau)$ at $\tau\leqslant t'$. If the duration τ of the ultrashort pulse is $\tau=8\tau_p$, then when $t'=\tau$, the integral (70) takes the form

$$I(\tau)=\frac{2\pi}{[1+2(\tau_p/\tau_D)^2]^{1/2}}\exp\left(-\frac{2\tau_p^2(\omega-\omega_{F_bF_a})^2}{1+2(\tau_p/\tau_D)^2}\right).\qquad(71)$$

The quantities τ_p, τ_D, γ_a, and γ_b in Eqs. (70) and (71) are unrelated by any constraint, and this makes it possible to examine both homogeneously broadened ($\gamma_{ba}\gg ku$) and inhomogeneously broadened ($\gamma_{ba}\ll ku$) transitions at any offset from resonance $\omega-\omega_{F_bF_a}$. The exponential time dependence $\mu(t')$ in (69) therefore makes it possible to determine experimentally the relaxation constants γ_a and γ_b independently of the nature of the broadening of the resonance transition.

The properties of $\mu(t')$ in the region $\tau\leqslant t'$, where the optical pumping effect shows up clearly in the photoinduced magnetization for any ratios of the spectral width Δ_ω, Doppler width ku, and relaxation constants γ, γ_a, and γ_b, can be investigated with the help of (69) if the inequalities (15) and (65) hold. In this case it is of interest to use an ultrashort pulse with left- hand circular polarization (61) in a tenuous gas

$$\gamma_a=0,\quad \gamma_b=\gamma,\quad \gamma_{ba}=\gamma/2.\qquad(72)$$

It is logical to look at the system after a long time

$$1\ll\gamma_b t',\qquad(73)$$

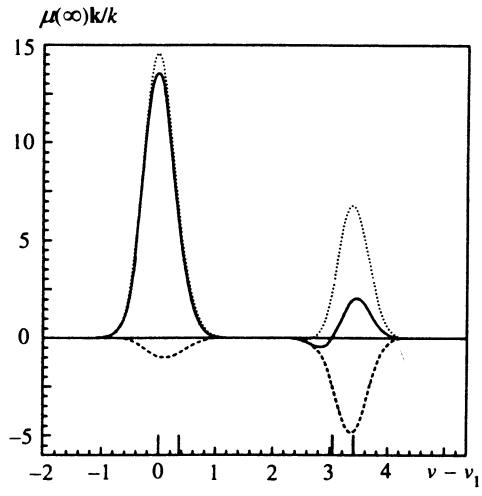

FIG. 3. Projection $\mu(\infty)\cdot\mathbf{k}/k$ of the constant photoinduced magnetization (74) as a function of $\nu-\nu_1$. The vertical unit is $5M_2/27$. The horizontal scale is the same as in Fig. 2, as is the sense of the dotted, dashed, and solid curves.

where the projection of the photoinduced magnetization (69) on the direction of the wave vector $\mathbf{k}$ assumes the constant asymptotic value

$$\mu(\infty)=M_2\sum_{F_bF_a}(2F_a+1)(2F_b+1)\begin{Bmatrix}J_b & F_b & I\\ F_a & J_a & 1\end{Bmatrix}^2$$
$$\times\left[\tilde g_{F_a}Q_a(F_a,F_b)+\sum_{F_a'}\tilde g_{F_a'}Q(F_a,F_a',F_b)\right]$$
$$\times\exp\left(-\frac{2\tau_p^2(\omega-\omega_{F_bF_a})^2}{1+2(\tau_p/\tau_D)^2}\right),\qquad(74)$$

where $\mu(\infty)=\mu(\infty)\mathbf{k}/k$,

$$M_2=\frac{2\pi M_0}{[1+2(\tau_p/\tau_D)^2]^{1/2}}.$$

Figure 3 shows $\mu(\infty)$ as a function of the center frequency $\nu=\omega/2\pi$ of the left-hand circular ultrashort pulse (61) for a gas containing atoms of $^{85}_{37}$Rb, for which Eqs. (57)–(63) hold; furthermore,

$$(E_{F_{a2}}-E_{F_{a1}})/h\gg ku/2\pi\sim\Delta_\omega/2\pi,\qquad(75)$$
$$(E_{F_{b2}}-E_{F_{b1}})/h\sim ku/2\pi\sim\Delta_\omega/2\pi.\qquad(76)$$

For the ultrashort pulse we assume

$$ku=1914\cdot10^6\mathrm{s}^{-1}(T=300\ K),\quad \Delta_\omega=3\cdot10^9\mathrm{s}^{-1},$$
$$\tau=8\tau_p=24\cdot10^{-10}\mathrm{s}.\qquad(77)$$

As follows from the relations (75) and (76), the four photoinduced magnetizations at the resonant frequencies $\nu=\nu_1$, $\nu=\nu_2$, $\nu=\nu_3$, and $\nu=\nu_4$ merge in pairs into two bell-shaped peaks, just as in the monochromatic wave displayed in Fig. 2. In addition, the first peaks in Figs. 2 and 3 are similar to one another, while the second peaks differ significantly. The latter difference stems from the fact that

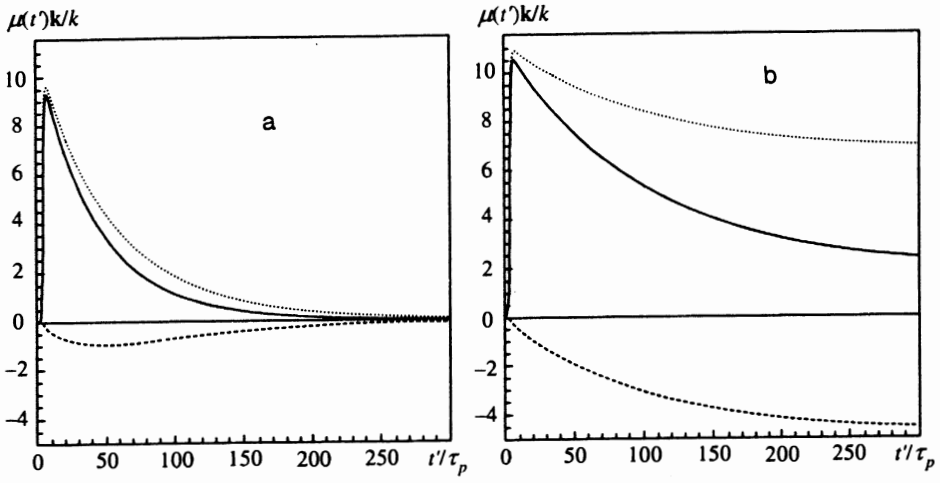

FIG. 4. Projection $\mu(t')\mathbf{k}/k$, of the photoinduced magnetization (69) as a function of t'/τ_p for $\nu=\nu_4$. The vertical unit is $5M_2/27$. The physical sense of the dotted, dashed, and solid curves is the same as in Fig. 2. a) $\gamma_a=1.5\gamma$ and $\gamma_b=2.5\gamma$; b) $\gamma_a=0$ and $\gamma_b=\gamma$.

the photoinduced magnetizations shown as dotted and dashed curves in Fig. 3 are similar in amplitude but of opposite sign, and they are shifted somewhat relative to one another. The total photoinduced magnetization near the transition frequencies ν_3 and ν_4 changes sign, as shown by the behavior of the solid curve. The differing behavior of the curves near $\nu=\nu_3$ and $\nu=\nu_4$ in Figs. 2 and 3 results mainly from the lack of a term with g_{F_b} in Eq. (74) to allow for the contribution of the resonant sublevels of the excited state E_b.

The behavior of the projection $\boldsymbol{\mu}(t')\cdot\mathbf{k}/k$ of the photoinduced magnetization (69) as a function of t'/τ_p is displayed in Figs. 4a and 4b for a gas containing $^{85}_{37}$Rb, with (57)–(63) and (75)–(77) holding for strict resonance $\nu=\nu_4$ at the transition frequency (60). However, in spite of the fact that $\nu=\nu_4$, by virtue of the Doppler effect (76) and optical pumping we find that all sublevels $E_{F_{a1}}$, $E_{F_{a2}}$, $E_{F_{b1}}$, and $E_{F_{b2}}$ participate in the magnetization of the atom. In Fig. 4a, because of collisions with active and impurity atoms, we set

$$\gamma_a=1.5\gamma, \quad \gamma_b=2.5\gamma, \quad \gamma_{ba}=2\gamma. \tag{78}$$

The photoinduced magnetization (69) can therefore be separated into three different parts. As a result of the resonant interaction of the atom with the ultrashort pulse, the first and second parts, with the factors $Q_a(F_a,F_b)\exp(-\gamma_a t')$ and $Q_b(F_b,F_a)\exp(-\gamma_b t')$, increase with time from zero to the maximum value at $t'_{m_1}\approx\tau$ and then decrease by virtue of the indicated exponentials, which describe the irreversible relaxation (dotted curve). Likewise, the third part, with the factor γ, starts to grow appreciably with time after the passage of the ultrashort pulse when $\tau<t'$, since it is due to the arrival of atoms in the sublevels $E_{F_{a1}}$ and $E_{F_{a2}}$ as a result of the spontaneous decay of the excited states of the atom with energies $E_{F_{b1}}$ and $E_{F_{b2}}$ (optical pumping effect). This third part has the opposite sign, and after the minimum value is reached at time

$$t'_{m2}=\frac{1}{\gamma_b-\gamma_a}\ln\frac{\gamma_b}{\gamma_a} \tag{79}$$

it increases to zero because of the difference of the exponentials $\exp(-\gamma_a t')-\exp(-\gamma_b t')$, as described by the dashed curve. In addition, the first maximum at $t'=t'_{m_1}$ is a sharp one by virtue of the fact that $\tau\ll 1/\gamma_a$ and $\tau\ll 1/\gamma_b$, while this bell- shaped maximum is so wide that when the three indicated parts are added, the total photoinduced magnetization in the form of the solid curve in Fig. 4a is strongly smoothed near the time $t'=t'_{m_2}$ of the second maximum. Here the presence of optical pumping leads to a more rapid decrease in the total photoinduced magnetization (solid curve), including contributions from all sublevels E_{a1}, E_{a2}, E_{b1}, and E_{b2}, than is observed in the absence of such pumping (dotted curve).

Equations (72) hold in a highly tenuous gas, and the time (79) of the second maximum becomes infinite. Ultimately [see (73)], then, the excited level E_b is completely emptied and the photoinduced magnetizations in the ground-state sublevels $E_{F_{a1}}$ and $E_{F_{a2}}$ become constant. The magnetization of the atom in these sublevels results both from the resonant interaction of the atom with an ultrashort pulse (dotted curve in Fig. 4b), and from optical pumping (dashed curve in Fig. 4b).

Experimentally, one usually investigates the electromotive force U of the receiver coil given by the well-known formula

$$U=-C_0\frac{\mathbf{k}}{k}\frac{d\boldsymbol{\mu}(t')}{dt'}, \tag{80}$$

where C_0 is a constant that depends on the experimental setup, and $d\boldsymbol{\mu}(t')/dt'$ is the time derivative of the photoinduced magnetization (69). We apply Eq. (80) to a gas containing active $^{85}_{37}$ Rb atoms with doublet splitting (62) in the ground state E_a and excited state E_b. A left-hand circular ultrashort pulse with center frequency $\nu=\omega/2\pi$ is characterized by Eqs. (75)–(77) and propagates in the gas with relaxation given by (78). Figure 5 shows the time variability of the emf U for various values of $\nu-\nu_1$; it is characterized by two peaks, which form as time increase in the range $0\leqslant t'\leqslant\tau$ and as the center frequency ν varies near the resonances $\nu=\nu_1$ and $\nu=\nu_2$ or $\nu=\nu_3$ and $\nu=\nu_4$. These peaks reflect the behavior of the integral (70) as a function of t' and $\nu-\nu_1$, and depend heavily on the profile of the ultrashort pulse. After the pulse has passed, with $\tau\leqslant t'$, U

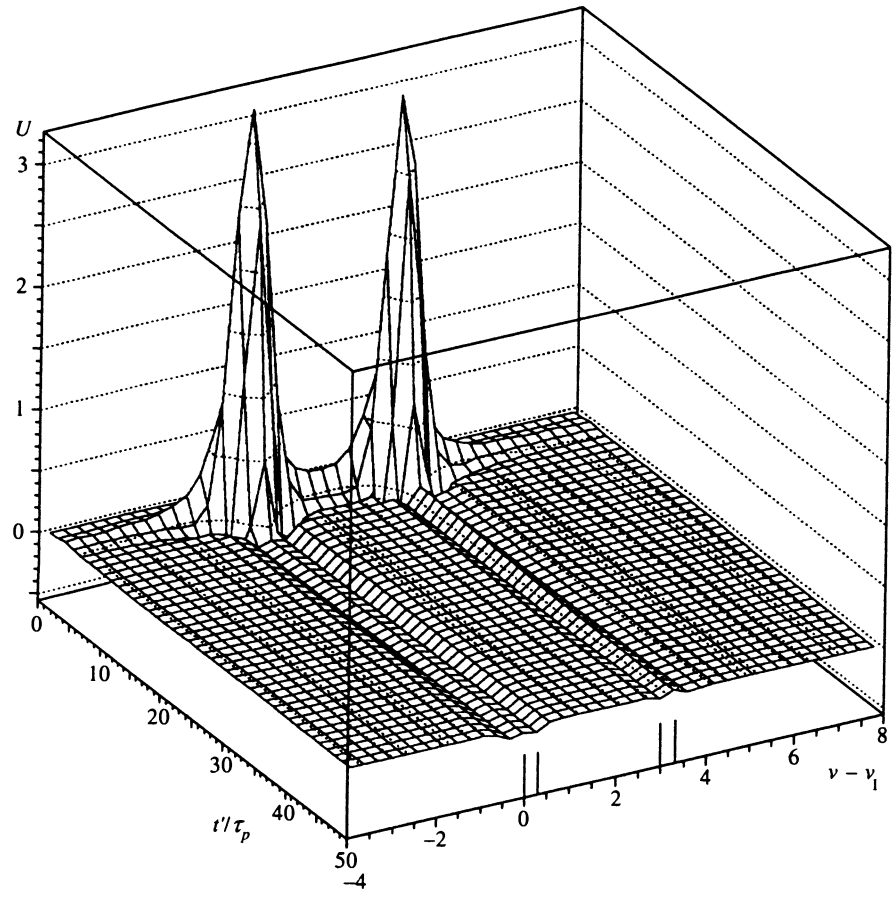

FIG. 5. Electromotive force U as a function of $\nu-\nu_1$ and t'/τ_p. The vertical unit is $-5C_0M_2/27\tau_p$; the horizontal unit is 1 GHz. The four long bars on the abscissa mark transition frequencies ν_1, ν_2, ν_3, and ν_4.

changes sign, and its behavior is governed by the relaxation described by the exponentials $\exp(-\gamma_a t')$ and $\exp(-\gamma_b t')$. These exponentials ensure that U vanishes in the limit $t'\to\infty$ (Fig. 6).

For small hyperfine splittings (44) in the excited state and large hyperfine splittings in the ground state $|\Delta_{F_a+1}-\Delta_{F_a}|\gg\Delta_\omega$, the magnetization induced by an ultrashort pulse at $0\leqslant t'$ can be obtained from Eqs. (46) and (64) in the form

$$\mu(t')=-\frac{\mathbf{k}}{k}\lambda\beta\sin(2\psi)M_0\sum_{F_aF_b}I(t')\times\left\{\left|\frac{d_{F_bF_a}}{d_{ba}}\right|^2\left[\tilde g_{F_a}Q_a(F_a,F_b)\times\exp(-\gamma_a t')+\frac{g_{J_b}}{g_{J_a}}\tilde g_{F_b}Q_b(F_b,F_a)\times\exp(-\gamma_b t')\right]+\frac{\gamma}{\gamma_b-\gamma_a}[\exp(-\gamma_a t')-\exp(-\gamma_b t')]\sum_{F_a'F_b'}\tilde g_{F_a'}Q(F_a,F_a',F_b,F_b')\right\}. \quad (81)$$

If the spectral width Δ_ω and the Doppler width ku are only small compared to the hyperfine splitting in the ground state

$$|\Delta_{F_a+1}-\Delta_{F_a}|\gg\Delta_\omega,ku,$$

then the arguments presented in connection with the inequality (43) still hold for Eq. (81).

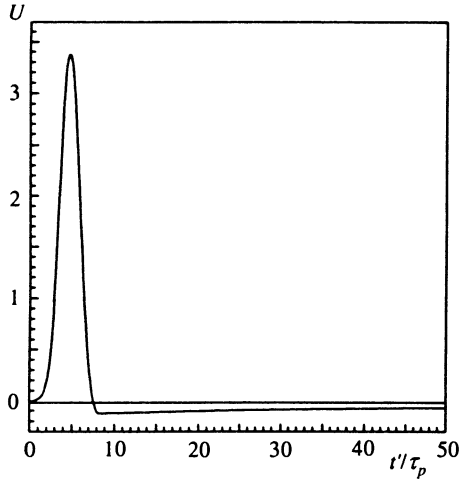

FIG. 6. Electromotive force U as a function of t'/τ_p for $\nu=\nu_4$. The vertical unit is $-5C_0M_2/27\tau_p$.

8. DISCUSSION

The dependence of the photoinduced magnetization in Eqs. (69) and (74) on the center frequency of an ultrashort pulse is to be interpreted in the sense that each individual pulse with center frequency $\nu=\omega/2\pi$ traverses the gas with the same initial conditions (8). If several such pulses traverse the gas in succession as the center frequency ν is swept, then each successive pulse—with shifted center frequency—must be delayed long enough with respect to its predecessor that relaxation can ensure that the equilibrium state dictated by the initial condition (8) is established in the gas. Otherwise, successive pulses will see initial conditions imposed by the optical polarization of the atoms produced by their predecessors, and Eqs. (69) and (74) will need to be refined.

When a light wave (1) with a stepped amplitude (47) traverses a gas with sufficiently strong collisions ($\gamma_a \sim \gamma_b$), the steady state (53), (55), and (56) is always established after the finite time interval (52). This means that the frequency of the light wave (1) can be experimentally swept so slowly that there is no noticeable departure from the steady state, and Eqs. (53), (55), and (56) retain their validity.

For gas atoms with zero nuclear spin in a strong magnetic field **H** in the Paschen–Back regime, the separation of the Zeeman sublevels of the split ground state E_a may be so large that only one such sublevel will be in resonance with the light wave (1), and optical pumping of nonresonant Zeeman sublevels of the same level will come into play. The optical pumping effect in photoinduced magnetization will then show up in the presence of a strong constant magnetic field **H**. As a result, certain properties of photoinduced magnetization[10] in a weak field for atoms with zero nuclear spin will be supplemented by new properties associated with optical pumping in a strong field.

Reference 6 shows the experimentally observed time-averaged plot of the emf (80) in a receiver coil, with the linearly swept frequency of the polarized light pulse near the $\lambda=7800$ Å $E_a \rightarrow E_b$ ($5^2S_{1/2} \rightarrow 5^2P^0_{3/2}$) transition of rubidium. The pulse duration $\tau=25\cdot 10^{-9}$ sec did not satisfy the inequality (15). The frequency separation obtained in the experiment of Ref. 6 between the two bell-shaped peaks of the emf is two orders of magnitude greater than the doublet splitting of the ground state of rubidium, and is therefore not associated with the hyperfine splitting of the levels. In addition, the magnetization of an isotropic gas by a linearly polarized light pulse observed in Ref. 6 is not consistent with symmetry, and it is also at variance with the results of the present paper. Thus, the behavior of the emf and magnetization of an isotropic gas by a linearly polarized light pulse in Ref. 6 require further investigation.

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